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Comment on acp-2022-400

Anonymous Referee #4

Referee comment on "Long-term declines in atmospheric nitrogen and sulfur deposition reduce critical loads exceedances at multiple Canadian rural sites, 2000–2018" by Irene Cheng et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-400-RC4>, 2022

This study presents an analysis of sulfur and nitrogen deposition monitored at 15 rural CAPMoN sites for the 2000-2018 time-period to characterize the spatiotemporal trends in atmospheric deposition across Canada and assess likely changes in ecosystem damage through inferences in changes in critical load exceedance during the period analyzed. The changes in atmospheric S and N deposition and the relative proportions of wet and dry deposition amounts reported by this analysis are along expected lines given the changes in $\text{NO}_x/\text{SO}_x/\text{NH}_3$ emissions and those reported in several similar analyses (both measurement and model based) for atmospheric deposition across the Continental U.S. Nevertheless, the documentation of these changes across Canada and from the CAPMoN measurements is a useful contribution. Before the manuscript can be considered suitable for publication, I feel there are aspects of the analysis that would benefit from additional elaboration as well as several conclusions that need more substantiation. The following comments are offered, addressing which may help improve the usefulness of this manuscript.

- From a total N deposition perspective, one of the shortcomings of this analysis relates to the limitations in the estimates of dry deposition amounts: (1) ambient measurements of select N species are combined with a deposition model to estimate their dry-deposition fluxes; (2) air sampling at majority of the locations only report HNO_3 , pNO_3 , and pNH_4 . The lack of measurements of NO_2 (possibly) and NH_3 (likely) represent a potentially large proportion of the dry deposited N and total atmospheric deposition budget. This largely renders the comparisons of wet vs dry N deposition amounts somewhat qualitative and confounds any quantitative interpretation of the relative contribution of oxidized vs reduced N to the total atmospheric N deposition amounts and their influence on subsequent CL exceedance and changes. To the authors credit, they do present the possible impact of these missing pieces through analyses of a more complete measurement set at the EGB site, which suggests non-negligible

impacts in terms of overall budget. It is not readily apparent whether the relative contributions of the missing components inferred from the EGB site could be extrapolated to the other locations, but perhaps a more explicit recognition of the missing N-deposition components in the discussion of results at other locations may help caveat the results more appropriately. As examples:

- L286: would dry deposition of N decrease by 50% if the estimates also included changes induced by NH₃ emissions?
 - L303-304: conceivably these contributions of HNO₃, pNH₄ and pNH₃ to dry N deposition would be different if NH₃ dry deposition was included. The particulate bound N dry deposition contributions could in fact be much smaller?
 - L306: It is possible that pNO₃ decreased in response to HNO₃ decreases because there was sufficient NH₃ available. The relative amounts of oxidized N to total N deposition could in fact be different if the atmospheric NH_x budget increased and that became a larger fraction of the N dry deposition. Should probably caveat this discussion by this important missing component.
 - L305: how representative is the fraction of non-routinely monitored N-species to the dry deposition budget (70%) at EGB of other locations?
 - L476-479: would the oxidized N deposition be greater than that of reduced N if NH₃ dry deposition estimates were also available? Perhaps this conclusion should be caveated since a potentially large contributor to the reduced N deposition is not accounted for. It is possible that some of the sites in Table 5 are not in proximity to NH₃ sources so that NH₃ dry deposition may not influence the conclusion – if that is the case it would be helpful to state that more explicitly.
 - L551: could the lack of trend in total deposition of reduced N be due to missing dry deposition of NH₃?
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- L70: the large variations in the contributions of dry deposition to total N-deposition across the U.S. convey significant spatial variability that likely arises from a combination of effects associated with spatial heterogeneity in emission source distribution, land-use and precipitation – it may be useful to state that explicitly.
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- L99: Is there a formal way to assess whether a site is “regionally representative” or is it just based on the rural settings of the site?
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- Equations 1-5: while a reference to the work of Zhang et al. (2008) is provided for the

- L415: what is the significance of comparing the annual rate of decrease in S and N dry deposition? Should also probably qualify this statement to indicate that the rate of decrease is expressed as percent.

- L470: I agree that quantifying the contribution of NO₂ and NH₃ dry deposition is important and that the current suite of measurements do not allow for their quantification. What other methods may be available to fill these critical data gaps? Can modeled deposition estimates of different species be used to fill in the gaps in the interim – are such estimates available from models such as GEM-MACH?

- L474: What does comparing the deposition fluxes of N and S tell us? Is there an ecological exposure significance of the relative mass fluxes of S and N?

- L638-640: Please elaborate why emission reductions were more effective in decreasing dry N deposition than wet N deposition? Some discussion of possible reasons leading to the noted "super-linear" (>100%) response in dry N deposition to emission reductions in the southeastern US should also be provided. Do contributions from transboundary transport play a role here? If so, which species would cause such behavior since HNO₃ deposits readily and would not be expected to undergo long-range transport?

- L659-660: It is not clear to me how the CL for the lakes were estimated - brief description or references to other work would be useful. Please also provide some discussion of what the max and min values of CL in Table S10 represent and how to

interpret these in context of the discussion in L684-697.

- L731: The sentence "the critical load analysis presented in this paper is a case study" is vague? Should a case study not be representative of conditions? Reasons for continued acidic state of lakes in eastern Canada should be mentioned briefly.